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# Phonon limit to simultaneous near-unity efficiency and indistinguishability in semiconductor single photon sources

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Semiconductor quantum dots have recently emerged as a leading platform to efficiently generate highly indistinguishable photons, and this work addresses the timely question of how good these solid-state sources can ultimately be. We establish the crucial role of lattice relaxation in these systems in giving rise to trade-offs between indistinguishability and efficiency. We analyse the two source architectures most commonly employed: a quantum dot embedded in a waveguide and a quantum dot coupled to an optical cavity. For waveguides, we demonstrate that the broadband Purcell effect results in a simple inverse relationship, where indistinguishability and efficiency cannot be simultaneously increased. For cavities, the frequency selectivity of the Purcell enhancement results in a more subtle trade-off, where indistinguishability and efficiency can be simultaneously increased, though by the same mechanism not arbitrarily, limiting a source with near-unity indistinguishability ( $> 99\%$ ) to an efficiency of approximately 96% for realistic parameters.

The efficient generation of on-demand highly indistinguishable photons remains a barrier to the scalability of a number of photonic quantum technologies<sup>1–4</sup>. To this end, attention has recently turned towards solid-state systems, and in particular semiconductor quantum dots (QDs)<sup>5–13</sup>, which can not only emit a single photon with high quantum efficiency, but can be easily integrated into larger photonic structures<sup>14</sup>, resulting in photons being emitted into a well-defined mode and direction. Highly directional emission is crucial to the overall efficiency of the source, and is typically achieved by either placing the QD in a waveguide with low out-of-plane scattering<sup>15,16</sup>, or by coupling resonantly to an optical cavity mode<sup>6–9,12,13</sup>. Nevertheless, the solid-state nature of QDs leads to strong coupling between the electronic degrees of freedom and their local environment; fluctuating charges<sup>17</sup>, nuclear spins<sup>18,19</sup>, and lattice vibrations<sup>20–23</sup> all lead to a suppression of photon coherence and a resulting reduction in indistinguishability<sup>11,24–27</sup>. While early experiments were indeed limited by these factors<sup>6–9</sup>, improvements in fabrication and resonant excitation techniques have steadily increased photon indistinguishability to levels now exceeding 99% in resonantly coupled QD–cavity systems<sup>12,13</sup>. Photon extraction efficiencies have also steadily improved, with the highest values reaching 98% in a photonic crystal waveguide<sup>16</sup>.

Despite this impressive progress, a system boasting very high ( $> 99\%$ ) indistinguishability and efficiency as required for e.g. cluster state quantum computing<sup>28</sup> remains elusive. Strategies aimed at achieving such a source typically focus on engineering the photonic environment in order to maximise the Purcell effect<sup>29,30</sup>, where the QD emission rate becomes  $F_P\Gamma$ , with  $\Gamma$  the bulk emission rate and  $F_P$  the Purcell factor<sup>29</sup>. Modelling a QD as a simple two-level-system with a Markovian phenomenological dephasing rate  $\gamma$ , the Purcell factor allows one to quantify the indistinguishability and efficiency as  $\mathcal{I} = \Gamma F_P / (\Gamma F_P + 2\gamma)$  and  $\eta = F_P / (F_P + 1)$

respectively<sup>31,32</sup>. In this simplistic model, one concludes that the Purcell factor is the key quantity of interest, which when increased will simultaneously lead to greater indistinguishability and efficiency.

In this work we demonstrate that this reasoning fails when one considers the coupling of the QD to its solid-state lattice at a microscopic level. We show that even in an idealised scenario, in which all other sources of noise are suppressed, the unavoidable coupling to phonons means neither waveguide nor cavity based sources can simultaneously reach near-unity indistinguishability and efficiency through Purcell enhancement alone.

## RESULTS

In contrast to simply introducing a Markovian dephasing rate, exciton–phonon coupling in the QD causes the lattice to adopt different configurations depending on whether the QD is in its ground or excited state [see Fig. 1]. As such, an excited to ground state transition accompanied by photon emission into the zero phonon line (ZPL) has a probability which scales as the square of the Franck–Condon factor  $B < 1$ , corresponding to the overlap of the two lattice configurations. The remaining emission events also scatter phonons in the process, resulting in emission of distinguishable photons, and a phonon sideband (SB) in the spectrum which must be removed. Due to the broadband nature of the Purcell enhancement in waveguides, the SB can only be removed by filtering. This necessarily sacrifices efficiency, resulting in a simple trade-off between indistinguishability and efficiency. For an emitter embedded in a moderate to high Q-cavity the phonon sideband can be naturally suppressed, though in this case the efficiency becomes  $\eta = B^2 F_P / (B^2 F_P + 1)$ , showing that removal of the sideband reduces the expected efficiency through the

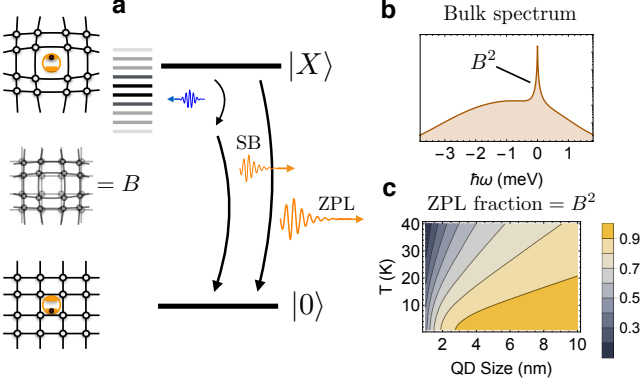


FIG. 1. Lattice relaxation in quantum dots and the phonon sideband. a, The ground state  $|0\rangle$  and single exciton state  $|X\rangle$  of a quantum dot are each associated with a different lattice configuration, shown on the left. b, Probability of emission into the zero phonon line (ZPL) scales as the square of the Franck–Condon factor  $B$  (lattice wavefunction overlap), with the remaining emission events constituting the phonon sideband (SB). c, The fraction of emission into the ZPL decreases with temperature and increases with quantum dot size.

Franck–Condon factor. This can in part be compensated by increasing the Purcell enhancement, though not indefinitely, as both the efficiency and indistinguishability drop when the strong coupling regime is reached. Based on a rigorous non-Markovian phonon theory, we derive analytic results quantifying the performance of single-photon sources for different architectures and in different regimes of operation.

### Phonon interactions in optically active QDs

The two key quantities used to characterise a single photon source are the efficiency, defined as<sup>31,33</sup>

$$\eta = \frac{P_D}{P_D + P_O}, \quad (1)$$

and the photon indistinguishability, defined as

$$\mathcal{I} = P_D^{-2} \int_{-\infty}^{\infty} d\omega \int_{-\infty}^{\infty} d\nu |S_D(\omega, \nu)|^2, \quad (2)$$

where the  $D$  and  $O$  subscripts denote the detected field and the field lost into unwanted modes. Here  $S_{D,O}(\omega, \nu) = \langle E_{D,O}^\dagger(\omega) E_{D,O}(\nu) \rangle$  is the generalised two-colour spectrum, with  $E_{D,O}(\omega)$  the positive component of the electric field in frequency space. Eq. (2) is more commonly (and equivalently) written<sup>31</sup>  $\mathcal{I} = P_D^{-2} \int_0^\infty dt \int_0^\infty d\tau |\langle \tilde{E}^\dagger(t + \tau) \tilde{E}(t) \rangle|^2$ , where  $\tilde{E}_{D,O}(t) = \int_0^\infty dt e^{-i\omega t} E_{D,O}(\omega) / (2\pi)$ . For  $\omega = \nu$  the two-colour spectrum is the measured emission spectrum, and the power into each channel is  $P_{D,O} = \int_{-\infty}^{\infty} d\omega S_{D,O}(\omega, \omega)$ . These expressions highlight the essential connection between the spectrum and performance of the source. We will analyse the three commonly used single photon

source architectures shown in Fig. 2 (a); a QD in a waveguide with Purcell enhancement (a slow-light waveguide) without (i) and with (ii) a spectral filter, and a QD coupled to a cavity (iii).

Calculation of the source figures of merit requires an accurate model of the dephasing processes affecting the QD. In addition to phonon induced processes, charge noise and spin noise can also affect emitted photon coherence<sup>18,19</sup>. However, our purpose here is assess the ultimate limits of a QD based source, and note that charge and spin noise can be heavily suppressed in suitably engineered samples<sup>12,13</sup>, while coupling to phonons can ever be completely quenched, as even at  $T = 0$  K phonon emission can still take place. We therefore focus on phonon induced dephasing mechanisms, with the understanding that our numerical results correspond to best case scenarios. Nevertheless, due to the very fast timescale ( $\sim$  ps) associated with phonon relaxation compared to the other dephasing mechanisms mentioned above, charge and spin noise can be readily included within our formalism by the introduction of Markovian dephasing rates, and our analytical expressions will explicitly include these rates also.

Of the possible phonon interactions that can take place in QDs, coupling to longitudinal acoustic (LA) phonons via deformation potential coupling has been shown to dominate<sup>20,21</sup>. Aside from lattice relaxation as captured by the Franck–Condon factor mentioned above, above a certain temperature LA phonons can also induce virtual transitions to QD states beyond the lowest single exciton state, giving rise to an additional phonon mediated decoherence process quite different in nature to the real phonon transitions represented by the emission spectrum sideband<sup>34</sup>. These processes are expected to be heavily suppressed at low temperatures ( $T < 10$  K), and will therefore be neglected in what follows, though once again we note that their inclusion could be easily achieved owing to the drastically different timescales involved.

With these arguments in mind, we consider a QD as a two-level-system with ground state  $|0\rangle$  and single exciton state  $|X\rangle$  with energy  $\hbar\omega_X$ <sup>21,25,35–38</sup>. The QD is coupled to a phonon and photon environment, giving the Hamiltonian  $H = \hbar\omega_X |X\rangle\langle X| + H_I^{PH} + H_I^{EM} + H_E^{PH} + H_E^{EM}$ , where  $H_I^{PH}$  and  $H_E^{EM}$  describe the free evolution of the phonon and photonic environments. The term  $H_I^{EM}$  contains the electric field operators  $E_{D,O}(\omega)$  which determine the spectrum, and describes the interaction between the QD and its photonic environment. Coupling to LA phonons is captured by the terms<sup>21,35</sup>,  $H_I^{PH} = \hbar |X\rangle\langle X| \sum_k g_k (b_k^\dagger + b_k)$  and  $H_E^{PH} = \hbar \sum_k \nu_k b_k^\dagger b_k$ , where  $b_k$  ( $b_k^\dagger$ ) is the annihilation (creation) operator of the phonon mode with wavevector  $k$  and frequency  $\nu_k$ . This interaction captures the mechanical deformation of the lattice when an exciton is present in the QD [see Fig. (1)]. Despite the complexity of the QD–phonon interaction, the harmonic nature of the phonons means their interaction with the QD can be fully characterised by the phonon spectral density, which for a spherically symmetric QD with harmonic confinement potential can be written<sup>23,39</sup>

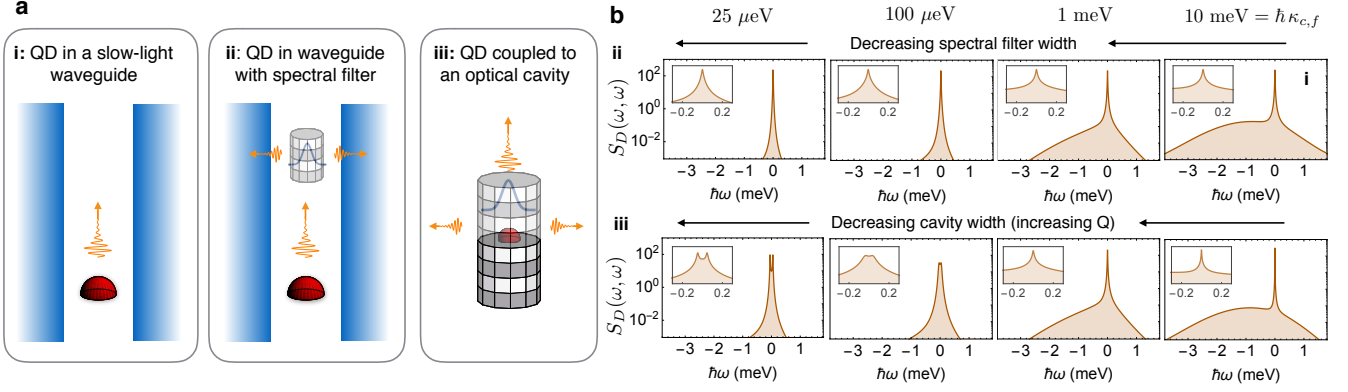


FIG. 2. Single photon source architectures and emission spectra. a, (i)–(iii) show the architectures we analyse: a QD emitting into a slow-light waveguide with and without a spectral filter, and a QD in a coherently coupled optical cavity. b, (ii)–(iii) show corresponding emission spectra as the filter or cavity is reduced in spectral width, demonstrating the filtering property of a cavity. The insets show a zoom-in of the ZPL features, highlighting ZPL broadening (Purcell enhancement) in the cavity case, which ultimately gives rise to vacuum Rabi splitting. The unfiltered spectrum for case (i) closely resembles the broad filter  $\hbar\kappa_f = 10$  meV case in (ii) as indicated. Parameters:  $T = 4$  K,  $\alpha = 0.03$  ps<sup>2</sup>,  $\hbar\xi = 1.45$  meV,  $\hbar\Gamma = 1$   $\mu$ eV; the waveguide in (i) and (ii) has Purcell factor  $\Gamma_D/\Gamma = 10$ , while the cavity in (iii) has  $\hbar g = 50$   $\mu$ eV, giving  $\Gamma_{\text{cav}}/\Gamma = 10$  when  $\hbar\kappa_c = 1$  meV.

$J_{\text{ph}}(\nu) = \sum_k |g_k|^2 \delta(\nu - \nu_k) = \alpha \nu^3 \exp[-\nu^2/\xi^2]$ . Here  $\alpha$  is an overall exciton–phonon coupling strength, and  $\xi = \sqrt{2}v/d$  is the phonon cut-off frequency, with  $v$  the speed of sound and  $d$  the confinement length (QD size). The cut-off frequency  $\xi$  defines a phonon energy scale above which interactions with the exciton are suppressed due to a mismatch in phonon and QD length scales.

Though the Hamiltonian given above, together with an appropriate choice of  $H_I^{EM}$  to model the relevant photonic environment, completely specifies the problem, calculating the two-colour spectra  $S_{D,O}(\omega, \nu)$  and by extension the source figures of merit is extremely challenging. In general the Hamiltonian is not easily diagonalised, and typically one therefore turns to approximate methods from the theory of open quantum systems, for example perturbative Markovian approaches such as the time-convolutionless master equation technique<sup>24,25</sup>. Since the emission spectrum sideband results from changes to the phonon environment (lattice relaxation), it is non-Markovian in nature<sup>40</sup>, and as such these Markovian treatments fail to capture it, yielding inaccurate source figures of merit<sup>24,25</sup>. Non-Markovian master equations can be employed<sup>25</sup>, though using these to calculate spectra requires extensions to the quantum regression theorem<sup>40</sup>, which had limited success when used to calculate photon indistinguishability, giving results that appeared not to approach the known analytic result in the limit of no cavity or filtering effects<sup>25</sup>. To date brute force numerical approaches, based on exact diagonalisation<sup>25,31</sup> or non-equilibrium Green’s functions techniques<sup>41</sup> have had the most success, though these provide limited insight into the underlying physical processes involved, and only in rare cases give analytic expressions.

To overcome these difficulties, we adopt a polaron transform approach, used in conjunction with formally solving the Heisenberg equations of motion for the emitted fields. This allows the dominant non-perturbative

non-Markovian phonon influence to be included, and permits us to derive analytic expressions in relevant regimes which elucidate the interplay between the Purcell and Franck–Condon factors, and trade-offs between efficiency and indistinguishability. Full details of the polaron transformation are given in the Supplementary information, though the central idea is to apply a displacement to the phonon mode operators dependent on the QD state,  $b_k \rightarrow b_k - |X\rangle\langle X| g_k/\nu_k$ , as this removes the original exciton–phonon coupling from the Hamiltonian<sup>23,38,42–44</sup>. Unitarity of the mode displacement means that the QD states must transform as  $|0\rangle \rightarrow |0\rangle$  and  $|X\rangle \rightarrow B_+|X\rangle$  with  $B_+ = \exp[\sum_k \nu_k^{-1} g_k (b_k^\dagger - b_k)]$ , and we can identify  $B_+$  as the operator achieving the necessary displacement of the lattice associated with the presence of an exciton. The Franck–Condon factor is then the thermal expectation value of this lattice displacement operator:

$$B = \langle B_+ \rangle = \exp \left[ -\frac{1}{2} \int_0^\infty d\nu \frac{J(\nu)}{\nu^2} \coth \left( \frac{\hbar \nu}{2k_B T} \right) \right]. \quad (3)$$

As mentioned, with no cavity or filtering effects only  $B^2$  of photon emission events go into the ZPL, with the remainder being incoherent in nature and constituting a phonon SB in emission spectra. As seen in Fig. 1 (b), while this phonon SB is orders of magnitude lower in intensity, its width is determined by the phonon cut-off frequency  $\hbar\xi \sim 1$  meV for typical parameters. As such, even at  $T = 0$  K where only phonon emission occurs, the sideband constitutes  $\approx 7\%$  of the emission, which increases with temperature and for QDs with smaller exciton localisation lengths, as seen in Fig. 1 (c).

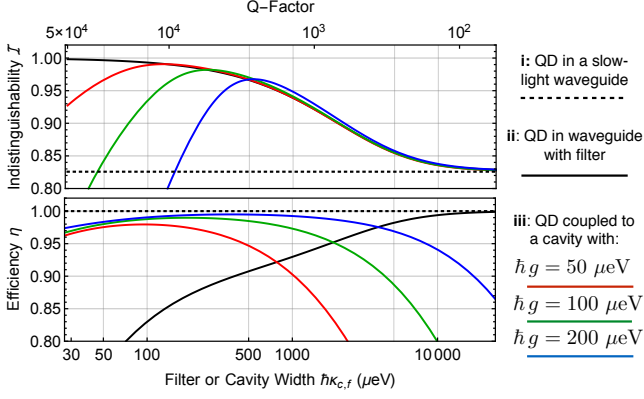


FIG. 3. Indistinguishability and efficiency of the three source setups shown in Fig. (2). The indistinguishability plot demonstrates that the dominant effect of a resonantly coupled cavity is to filter the QD emission, while the efficiency plot demonstrates that Purcell enhancement in a cavity can overcome efficiency losses incurred by filtration of the phonon sideband. Cavity Q-factors on the upper x-axis correspond to a cavity resonance  $\hbar\omega_c = 1.4$  eV. Parameters as in Fig. (2), giving  $B^4 = 83\%$ , and we have assumed a loss-less waveguide  $\Gamma_O = 0$  for (i) and (ii), while for (iii)  $\Gamma_O = \Gamma$ .

### Emission properties

Our task now is to understand how a spectral filter or cavity can affect the detected spectrum  $S_D(\omega, \nu)$ , which will in turn affect the indistinguishability via Eq. (2) by, for example, removing the phonon SB. Crucially, however, we also need to understand the quantitative relationship between the detected and lost (out-of-plane) spectrum  $S_O(\omega, \nu)$  when these filtering or cavity effects are introduced, since this will affect the source efficiency via Eq. (1).

As shown in Methods, the two-colour-spectra are found by solving the Heisenberg equations of motion for the electric field operators, and in all cases (i)–(iii) we find it is possible to write  $S_D(\omega, \nu) = \mathcal{G}(\omega, \nu)S_O(\omega, \nu)$ . The function  $\mathcal{G}(\omega, \nu)$  is a Green’s function, describing how the field is transformed propagating from its creation at the QD, to the detector. For the unfiltered waveguide source (i)  $\mathcal{G}(\omega, \nu) = \Gamma_D/\Gamma_O$ , with  $\Gamma_D$  and  $\Gamma_O$  the emission rates into and out of the waveguide, showing that the in-plane spectrum is simply a frequency independent enhancement of the out-of-plane spectrum. For the filtered waveguide source (ii)  $\mathcal{G}(\omega, \nu) = (\Gamma_D/\Gamma_O)h_f^*(\omega)h_f(\nu)$ , where  $h_f(\omega) = (\kappa_f/2)[i(\omega - \omega_f) - (\kappa_f/2)]^{-1}$  with  $\kappa_f$  and  $\omega_f$  the filter width and central frequency respectively. The filter now fundamentally changes the detected spectrum, as we might expect. As a key insight of this work, in case (iii) for the optical cavity we find  $\mathcal{G}(\omega, \nu) = (\Gamma_{\text{cav}}/\Gamma_O)h_c^*(\omega)h_c(\nu)$ , where now  $h_c(\omega) = i(\kappa_c/2)[i(\omega - \omega_c) - \kappa_c/2]^{-1}$  and  $\Gamma_{\text{cav}} = 4g^2/\kappa_c$ , with  $g$  the light-matter coupling strength,  $\kappa_c$  the cavity width, and  $\omega_c$  the cavity mode frequency.

Comparing cases (ii) and (iii) above, we see that there is a formal analogy between a spectral filter and an optical cavity, as has been alluded to elsewhere<sup>43,45</sup>. That

is not to say, however, that the two are equivalent; as a filter is reduced in width and the sideband removed, one simply moves photons from the detected channel to the out-of-plane channel. As a cavity is reduced in width, however, the strength of the light-matter coupling is modified, giving rise to Purcell enhancement of emission events resonant with the cavity, while also removing the sideband. Unlike a filter, this cavity enhancement can overcome sideband photons that are now being lost due to cavity filtering effects. The broadband nature of Purcell enhancement in waveguides means a waveguide with Purcell enhancement and a filter is not equivalent to a cavity, since in the former case both the ZPL and the sideband are enhanced. Detected spectra for the waveguide with filter (ii) and cavity (iii) are shown in Fig. 2 (b), where the waveguide has a Purcell factor of  $\Gamma_D/\Gamma = 10$ . In addition to filtering effects seen in both cases, the insets show that in the cavity case, frequency selectivity of the Purcell enhancement gives ZPL broadening, and ultimately signs of vacuum Rabi splitting as the strong coupling regime is reached.

### Waveguide vs Cavity Comparison

In Fig. (3) we compare the three single photon source architectures shown in Fig. 2 (a). For large cavity or filter widths ( $\kappa_{c,f} \gg \xi \sim 1$  meV/ $\hbar$ ), the entire sideband contributes to the detected field [see Fig. 2 (b)], yielding an indistinguishability of that in bulk,  $\mathcal{I} = B^4 \approx 83\%$  for realistic parameters at  $T = 4$  K. As the filter or cavity is reduced in width, the indistinguishability increases as the phonon sideband is removed. This plot demonstrates that until the strong coupling regime is reached, i.e. for  $\kappa_c > 4g$ , with regards to the indistinguishability, the dominant effect of the cavity is that of filtering, as also suggested by Fig. 2 (b). The efficiency of the filtered source (ii), however, always decreases with decreasing filter width as the sideband is removed, whereas the cavity efficiency (iii) increases, since the Purcell effect compensates for photons lost into the sideband.

To elucidate these points, let us consider the experimentally relevant regime where the filter or cavity width is larger than any features present in the ZPL. This corresponds to  $\Gamma_D < \kappa_f$  in case (ii), and  $\Gamma_{\text{cav}} < \kappa_c$  in case (iii), meaning that the strong coupling regime is not reached. In this regime we find that the master equation describing the QD degrees of freedom can be approximated as  $\dot{\rho} = \Gamma_{\text{tot}}\mathcal{L}_\sigma[\rho(t)] + 2\gamma_{\text{tot}}\mathcal{L}_{\sigma^\dagger\sigma}[\rho(t)]$ , where for (i) and (ii)  $\Gamma_{\text{tot}} = \Gamma_O + \Gamma_D$  and  $\gamma_{\text{tot}} = \gamma$ , and for case involving the cavity (iii)  $\Gamma_{\text{tot}} = \Gamma_O + \Gamma_{\text{cav}}$  and  $\gamma_{\text{tot}} = \gamma + \gamma_{\text{ph}}$  with  $\gamma_{\text{ph}} = 2\pi(gB/\kappa_c)^2 J_{\text{ph}}(2gB) \coth(\hbar gB/k_B T)$  a Markovian phonon-induced ZPL dephasing rate. We have introduced a phenomenological dephasing rate  $\gamma$  to capture e.g. charge or spin noise, which is a valid procedure provided it is uncorrelated with any phonon processes. With this master equation we find that the indistinguishability

can be approximated by

$$\mathcal{I} = \frac{\Gamma_{\text{tot}}}{\Gamma_{\text{tot}} + 2\gamma_{\text{tot}}} \left( \frac{B^2}{B^2 + \mathcal{F}[1 - B^2]} \right)^2, \quad (4)$$

where  $\mathcal{F} = \int_{-\infty}^{\infty} d\omega |h_{f,c}(\omega)|^2 S_{\text{SB}}(\omega, \omega) / \int_{-\infty}^{\infty} d\omega S_{\text{SB}}(\omega, \omega)$  is the fraction of the sideband not removed by the filter or optical cavity. The first factor in Eq. (4) is similar to the phenomenological expression<sup>31</sup>, though with an additional phonon-induced dephasing rate  $\gamma_{\text{ph}}$ . The second factor, however, highlights the essential role of the Franck-Condon factor  $B$ , and the interplay between this and the fraction of the sideband remaining in the spectrum  $\mathcal{F}$ . The efficiency in this regime is given by

$$\eta = \frac{\Gamma_{\text{cav}}(B^2 + \mathcal{F}[1 - B^2])}{\Gamma_{\text{cav}}(B^2 + \mathcal{F}[1 - B^2]) + \Gamma_O}, \quad (5)$$

for the cavity, and  $\eta = (B^2 + \mathcal{F}[1 - B^2])\Gamma_D/(\Gamma_D + \Gamma_O)$  for the waveguide, again demonstrating the importance of the Franck-Condon factor.

For a broad filter or low-Q cavity, for which  $\kappa_{f,c} \gg \xi \sim 1 \text{ meV}/\hbar$ , we have  $\mathcal{F} = 1$  and Eq. (4) becomes  $\mathcal{I} = B^4\Gamma_{\text{tot}}/(\Gamma_{\text{tot}} + 2\gamma_{\text{tot}})$ . Since  $B < 1$ , the phonon sideband reduces the indistinguishability that would be expected from Markovian or phenomenological treatments. The efficiencies in this regime become  $\eta = \Gamma_D/(\Gamma_D + \Gamma_O)$  in the waveguide case, while for the cavity we find  $\eta = \Gamma_{\text{cav}}/(\Gamma_{\text{cav}} + \Gamma_O)$ , becoming  $\eta = F_{\text{cav}}/(F_{\text{cav}} + 1)$  for  $\Gamma_O = \Gamma$  with  $F_{\text{cav}} = 4g^2/(\kappa_c\Gamma)$  the cavity Purcell factor. Thus, in this regime the efficiencies are equal to those expected from phenomenological approaches<sup>31</sup>.

For a sufficiently narrow filter or cavity, for which  $\kappa_{f,c} \ll \xi$ , we have  $\mathcal{F} \approx 0$ , and Eq. (4) becomes  $\mathcal{I} = \Gamma_{\text{tot}}/(\Gamma_{\text{tot}} + 2\gamma_{\text{tot}})$ . Here the cavity or filter removes the phonon sideband from the detected spectrum, increasing the indistinguishability as compared to that found for a broad filter or low-Q cavity. Although the sideband appears not to affect the indistinguishability of the source in this regime, the efficiency drops monotonically in case (ii), and for the cavity (iii) becomes  $\eta = B^2\Gamma_{\text{cav}}/(B^2\Gamma_{\text{cav}} + \Gamma_O)$ . Now we see the Franck-Condon factor acting to reduce the source efficiency<sup>31</sup>, which demonstrates a trade-off between the two source figures of merit. Crucially, however, the increase in  $\Gamma_{\text{cav}} = 4g^2/\kappa_c$  with decreasing cavity width  $\kappa_c$  can compensate for sideband photons which are lost, giving rise to an overall increase in efficiency as  $\kappa_c$  is reduced.

Considering lastly the strong coupling regime for the cavity case (iii), where  $4g > \kappa_c$ , we see from Fig. (3) that the indistinguishability begins to drop sharply, indicating that the cavity-based source cannot be arbitrarily improved by decreasing  $\kappa_c$  (or increasing  $g$ ). In this regime Rabi oscillations occur between the QD and cavity, allowing Markovian phonon-induced dephasing mechanisms to have a greater effect. Moreover, these Rabi oscillations give the excitation a greater probability to be lost to non-cavity modes, as seen by the corresponding drop in efficiency.

## DISCUSSION

Our results allow for a critical appraisal of the most commonly used single photon source architectures. For a QD in a perfect lossless waveguide, although efficiencies may well approach 1, even in the absence of pure-dephasing ( $\gamma = 0$ ), the broadband nature of Purcell enhancement means that the unavoidable phonon sideband in the emission spectrum limits photon indistinguishability to approximately  $B^4 = 83\%$  at  $T = 4 \text{ K}$ . A filter can improve this value, but the efficiency will then necessarily decrease, giving  $\mathcal{I} \approx 99\%$  and  $\eta = 83\%$  for a filter width of  $\hbar\kappa_f = 100 \text{ } \mu\text{eV}$ .

For a QD coupled to a cavity, we can identify an optimal regime where  $4g < \kappa_c \ll \xi$ , such that the cavity removes the sideband, but is not so narrow as to enter the strong coupling regime. Clearly a small QD-cavity coupling strength  $g$  most easily satisfies this criterion, though this comes at the expense of a reduced efficiency as the cavity Purcell effect weakens. These competing requirements mean a cavity-based source cannot simultaneously reach near-unity efficiency and indistinguishability by simply increasing the cavity Q-factor or QD-cavity coupling strength. Nevertheless, readily achievable experimental values of  $\hbar g = 30 \text{ } \mu\text{eV}$  and  $\hbar\kappa_c = 120 \text{ } \mu\text{eV}$  give  $\mathcal{I} = 99\%$  and  $\eta = 96\%$  at  $T = 4 \text{ K}$ .

These numbers and the calculations in Fig. (3) are based on a favourable but realistic scenario, in which phonons are the dominant source of dephasing, and placing the QD in a cavity does not affect its emission into non-cavity modes. This immediately points us towards how source architectures may be improved, as the figures of merit are ultimately limited by the size of the phonon sideband in the bulk QD spectrum and the strength of emission into non-cavity modes. The former may be reduced in QDs with a larger exciton localisation length<sup>21</sup>, or actively suppressed by manipulation of the phononic density of states. Both of these approaches, however, come at the risk of increasing ZPL dephasing<sup>46</sup> which must be avoided. Perhaps more promising is the prospect of decreasing photon emission into non-cavity modes. Our results suggest that future cavity designs ought to carefully take into account the spectrum and strength of emission into these leaky modes, as well as the usual cavity mode volume and Q factor. Decreased emission into non-cavity modes is possible for low Q-cavities<sup>47</sup>, though these cavities will not be spectrally narrow enough to remove the sideband. Instead, a photonic environment that strongly suppresses all emission except into a spectrally narrow ( $\sim 0.1 \text{ meV}$ ) cavity mode is required.

## METHODS

To find the detected and out-of-plane electric fields which determine the relevant emission properties we write  $E_\mu(\omega) = \sum_l c_{\mu,l}(\omega)$ , where  $c_{\mu,l}(\omega)$  is the annihilation operator for mode  $l$  of environment  $\mu$  moved into the Heisenberg picture and Fourier transformed, with  $\mu = \{D, O\}$  denoting the detected ( $D$ ) and out-of-plane



(O) channels. The way in which the mode operators  $c_{\mu,l}$  (and hence the fields) couple to the QD is contained within the Hamiltonian term  $H_f^{EM}$ , and depends on the source architecture under consideration, with the full details given in the Supplementary information. In all cases, equations of motion coupling the electric fields to the QD degrees of freedom are obtained from the polaron transformed Hamiltonian, and therefore contain bath displacement operators which give rise to a phonon sideband.

For case (i), a defining characteristic of slow-light waveguides is the broadband nature of the Purcell enhancement<sup>30</sup>. We therefore assume a flat photonic spectrum over frequencies relevant to the QD, from which we find the detected and out-of-plane fields are  $\tilde{E}_{D,O}(t) \approx i\sqrt{\Gamma_{D,O}/2\pi}\tilde{\sigma}(t)\tilde{B}_-(t)$  in the time-domain, where  $\Gamma_{D,O}$  is the corresponding emission rate,  $\sigma = |0\rangle\langle X|$ , and tildes indicate Heisenberg picture operators. The above expression has the same form as that of a standard quantum dipole emitter, though modified by a lattice displacement operator  $B_-$ , which through Eqs. (1) and (2) affects the spectrum, efficiency and indistinguishability. For case (ii), the effect of a spectral filter is most easily introduced in the frequency domain, where the detected field becomes  $E_D(\omega) = \sqrt{\Gamma_D/\Gamma_O}h_f(\omega)E_O(\omega)$ <sup>48</sup>, and for a Lorentzian filter we have  $h_f(\omega) = (\kappa_f/2)[i(\omega - \omega_f) - (\kappa_f/2)]^{-1}$  with  $\kappa_f$  and  $\omega_f$  the filter width and central frequency respectively. Introducing the filter in this way requires that we add a term  $(\Gamma_D/\Gamma_O)\int_{-\infty}^{\infty} d\omega[1 - |h_f(\omega)|^2]S_O(\omega, \omega)$  to the denominator in Eq. (1) to include the field rejected by the filter. In the time domain the detected field takes the form of a convolution between the emitted field and the filter response function.

We follow a similar procedure for case (iii), though now explicitly account for variation of the cavity line-shape across the relevant QD frequencies. The out-of-plane emission (i.e. not via the cavity mode) is given by  $\tilde{E}_O(t) \approx i\sqrt{\Gamma_O/2\pi}\tilde{\sigma}(t)\tilde{B}_-(t)$ , which takes the same form as in case (i). We make the assumption that the detected field consists of those photons emitted by the cavity mode<sup>24,27,41,45</sup>. Although it is customary to define the detected field in this way for QD-cavity systems, one expects that in the very broad cavity limit

the detected field will also contain a contribution arising from direct QD emission. We do not include this contribution in our calculations, though note that its effect would be to slightly raise efficiencies in the less interesting  $\kappa_c \gg 4g$  regime for case (iii). Taking the usual detected field definition, we find it can be written in frequency space as  $E_D(\omega) = \sqrt{4g^2/\kappa_c}\Gamma_O h_c(\omega)E_O(\omega)$ , with  $h_c(\omega) = i(\kappa_c/2)[i(\omega - \omega_c) - \kappa_c/2]^{-1}$ , where  $g$  is the light-matter coupling strength,  $\kappa_c$  the cavity width, and  $\omega_c$  the cavity mode frequency. Comparing to case (ii) above, this expression demonstrates the analogy between a cavity and a spectral filter, and the mathematical connection between filtering effects and the phonon sideband captured in the operator  $\tilde{B}_-(t)$ . One can see that coupling to a cavity has two dominant effects. The first is to modify the QD dynamics, which is captured implicitly in the time-dependence of the operator  $\tilde{\sigma}(t)$ . How these dynamics are modified will depend on the regime of light-matter coupling, and will include Purcell enhancement, as well as phonon induced dephasing mechanisms<sup>24</sup>. The second is to spectrally filter the resulting QD emission, as described by the cavity filter function  $h_c(\omega)$ . With these relationships between the electric fields, it follows that the spectra can be written  $S_D(\omega, \nu) = \mathcal{G}(\omega, \nu)S_O(\omega, \nu)$ .

Finally, we note that the relationship  $S_D(\omega, \nu) = \mathcal{G}(\omega, \nu)S_O(\omega, \nu)$  is exact in cases (i) and (ii). In case (iii) it is exact in the absence of coupling to phonons, valid in both the strong and weak QD-cavity coupling regimes. As discussed in detail in the supplementary information, when phonons are included, the theory remains quantitatively accurate except in the very strong coupling regime where dissipative terms in the master equation not included in the Green's function  $\mathcal{G}(\omega, \nu)$  become important. Nevertheless, in this regime the present theory remains qualitatively accurate when compared to an exact approach, and correctly predicts the fall in source merit criteria with decreasing cavity width.

### Data availability

The data that support the plots within this paper and other findings of this study are available from the corresponding author upon reasonable request.

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#### **AUTHOR CONTRIBUTIONS**

J.I.-S. and D.P.S.M. contributed equally to this work. All authors were involved in the conception, develop-

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#### **ADDITIONAL INFORMATION**

Supplementary material is available for this work. Correspondence and requests for materials should be addressed to J.I.-S.

#### **COMPETING FINANCIAL INTERESTS**

The authors declare no competing financial interests.